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Environmental impact of nanomaterials in composite membranes: Life cycle assessment of algal membrane photoreactor using polyvinylidene fluoride – composite membrane

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ABSTRACT

This study assessed the environmental impacts of a composite polyvinylidene fluoride (PVDF) membrane (incorporating nanomaterials) and compared with neat PVDF membrane on algal membrane photoreactor (A-MPR) system's overall sustainability. The life cycle assessment (LCA) was carried out using Simapro 8.4.0 with cradle-to-gate approach, including raw materials, equipment, transportation and electricity consumption using ReCiPe 1.13 (H) and IPCC 2013 GWP 100a methodology. From the LCA analysis, silver/graphene oxide - polyvinylidene fluoride (Ag/GO-PVDF) membrane fabrication showed higher environmental impact than the neat PVDF membrane fabrication due to the addition of Ag/GO nanohybrids into the polymer. However, the A-MPR system using the Ag/GO-PVDF membrane exhibited better environmental footprint due to the improved performance of the modified membrane in producing higher volume of permeate as the output. Therefore, the A-MPR system using Ag/GO-PVDF membrane had outweighed the additional environmental impact of the Ag/GO-PVDF membrane fabrication process. Energy demand was identified as the main environmental hotspot in the LCA analysis. Subsequently, sensitivity analysis was performed to find out the effect of various energy mix for electricity generation towards the environment. The analysis revealed that the energy source for electricity generation had significant influence on the overall sustainability of the A-MPR system. The use of grid with 100% renewable energy (hydropower and geothermal) and solar photovoltaic might be able to mitigate 94.8% and 97.5% of CO_2 emission, respectively.

1. Introduction

The development of nanoparticles and nanohybrids has been accelerating in recent years. Among the nanomaterials that have been extensively studied by researchers are ZnO, TiO_2 and Ag, and their incorporation with graphene oxide (GO) to form composite nanomaterials has gained tremendous interest. These nanomaterials possess excellent anti-microbial, anti-corrosive properties and high thermal stability (Chung et al., 2017). Therefore, they are widely used in biomedical, optics, electronics and gloves industries (Kołodziejczak-Radzimska and Jesionowski, 2014; Murphy et al., 2015; Wang et al., 2010). Various nanomaterials have been

Abbreviations: Climate change, CC; Ozone depletion, OD; Terrestrial acidification, TA; Freshwater eutrophication, FE; Marine eutrophication, MEP; Human toxicity, HT; Photochemical oxidant formation, POF; Particulate matter formation, POF; Terrestrial ecotoxicity, TET; Freshwater ecotoxicity, FET; Marine ecotoxicity, MET; Ionizing radiation, IR; Agricultural land occupation, ALO; Urban land occupation, ULO; Natural land transformation, NLT; Water depletion, WD; Metal depletion, MD; Fossil depletion, FD.

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incorporated into polymeric membranes to produce composite membranes with enhanced performance. These composite membranes could be produced by phase inversion (Chong et al., 2017a,b), layer-by-layer deposition (Zuin et al., 2013) or self-assembly methods (Teow et al., 2012). The incorporation of nanomaterials in membrane polymer could increase the membrane pore size and wettability while reducing membrane surface roughness. As a result, the permeability and antifouling properties of the membranes were enhanced (Chong et al., 2017a,b; Ho et al., 2017; Leo et al., 2012). Lee et al. (2013) reported membrane incorporated with GO performed 5 times greater than the neat PSF membrane in a membrane bioreactor (MBR) filtration process. Kumar et al. (2016) has also proven that GO-TiO_2 incorporated membrane increased pure water flux by 28%, and at the same time increased humic acid rejection from 80.4% to 97.0%.

PVDF polymer has high thermal stability and chemical resistance due to its high carbon-fluoride dissociation energy. Therefore, it is widely used in the membrane technology especially for alkaline or acidic wastewater separation application (Yeow et al., 2004). However, as PVDF is a semi-crystalline polymer, it is relatively hydrophobic than other polymers like polyethersulfone (PES) and polysulfone (PSF). As hydrophobic membrane always shows low permeation flux and low anti-fouling properties in wastewater treatment application, many studies have been performed to modify the PVDF membrane to enhance its hydrophilicity for higher throughput and better fouling resistance. Bae and Tak (2005) attempted to incorporate TiO_2 nanoparticles in PVDF membranes and found that the pure water flux increased by 9.2%. In another study by Zhao et al. (2014) reported that GO embedded PVDF membrane was able to increase the critical flux up to 50% compared with the commercial PVDF membrane in a MBR system. Zhang et al. (2015) had successfully increased hydrophilicity, membrane negative charge and mechanical strength of PVDF membrane, which lead to enhancement of the membrane anti-fouling ability via the addition of TiO_2/polyethylene glycol.

To date, algal membrane photoreactor (A-MPR) has been identified to be a green, sustainable system that has great potential to be scale-up for biofuel generation purpose. Integrating wastewater treatment and microalgae cultivation enable wastewater to be polished via biological nutrient up-take by microalgae and simultaneously harvesting microalgal biomass for biofuel or other valuable products generation (Cheah et al., 2016; Menetrez, 2012). However, membrane fouling is always an obstacle for smooth operation of the system. Membrane fouling in the A-MPR system will trigger frequent backwash, resulting in excessive use of cleaning chemical and reduce the lifespan of the membrane. The employment of GO based composite membranes appear to be a fine solution to reduce the fouling propensity and subsequently reduce the membrane backwash frequency. However, the impacts of adding GO nanomaterials into the membrane toward the environment and the sustainability of the A-MPR system are unknown. The benefit of the application shall be considered against the potential impacts towards the environment and human health, which can be further evaluated systematically using life cycle assessment (LCA) approach.

LCA is a process which evaluates the impact of a product starting from its raw material extraction to each production step until its end-of-life towards the environment and human health by accumulating a set of inventory which is relevant to the inputs and outputs of the objects studied (Suhariyanto et al., 2017; Zuin et al., 2013). One of the latest LCA methodology is ReCiPe, which consists of two approaches: CML and Ecoindicator 99. The formal is a midpoint indicator and the latter is an endpoint indicator. To date, there are some LCA studies reported for nano-based products and only a few on nano-technological production methods. The environmental impact of TiO_2 composite membrane using layer-by-layer technology was investigated by Zuin et al. (2013). The deposition of TiO_2 nanoparticles by electrostatic method on PES membrane showed insignificant effect on all of the impact categories studied. Instead, the major impact contributors in the making of PES membranes were the solvent, polymer and electricity used. Stieberova et al. (2017) assessed the environmental benefits of nanoparticles incorporated coating to produce self-cleaning metal panels. The LCA results indicated that the cumulative energy demand (CED) value of the PVDF was significantly lower than the treated ZnO nanoparticles in their manufacturing process. However, during the use stage, the ZnO nanoparticles incorporated PVDF coating which was maintenance-free showed better performance in all assessed impact categories throughout its whole life cycle.

Our previous study found that PVDF membrane with Ag/GO nanohybrids showed better anti-fouling ability and anti-biofouling potential compared with GO nanosheets and ZnO/GO nanohybrids in an algal organic matter filtration study (Chong et al., 2017b). Although the use of GO nanomaterials in membrane application has been studied extensively through experimental work, the potential impacts of the addition of these materials towards environment and human health have not been widely investigated. To the best of our knowledge, no LCA study has been performed to investigate the impacts of GO based composite membrane in the A-MPR. Hence, this study aims to investigate the benefit of using the Ag/GO-PVDF membrane in comparison with neat PVDF membrane in an A-MPR application in order to identify the most environmental-friendly setup. The impacts were evaluated using cradle-to-gate approach by ReCiPe and IPCC. The effects on all the impact categories in membrane fabrication stage and A-MPR operation stage were evaluated in details and further recommendations were made to present a product with high degree of environmental friendliness.

2. Methodology

2.1. Process description

In this study, the membranes and A-MPR system are the major elements for the life cycle assessment. Hence, the preparation process of neat PVDF or Ag/GO-PVDF membrane and the A-MPR setup procedures were briefly described in this section.

2.1.1. Membrane fabrication

Prior to membrane synthesis, the Ag/GO nanohybrid was prepared as per described in the previous study by (Mahmoudi et al., 2015). Firstly, GO suspension was mixed with aqueous silver nitrate ($AgNO_3$) solution. Then, sodium borohydride ($NaBH_4$) solution was added slowly into the $AgNO_3$-GO suspension to reduce the $AgNO_3$ and form Ag nanoparticles. From the results, the Ag nanoparticles were distributed evenly across the GO nanosheets with an average size of 2–5 nm. On the other hand, the fabrication of neat PVDF or Ag/GO-PVDF membrane were performed via phase inversion process. The PVDF concentration was fixed at 18 wt% with the addition of 0.1 wt% of PVP dissolved in DMAc solvent, while 0.4 wt% of Ag/GO nanohybrid was added for the preparation of composite membrane. Further details and structural characterizations could be found in our previous study of AOM filtration with PVDF-GO nanohybrid membranes (Chong et al., 2017b).

2.1.2. A-MPR system

Microalgae *Chlorella Vulgaris* (*C. Vulgaris*) with cell density of 40×10^6 cell/mL was cultivated in the A-MPR. Municipal synthetic wastewater was treated in the A-MPR tank with a retention time of

5 d. The quality of feed water is shown in Table A1. The A-MPR setup is presented in Fig. 1. Air was supplied at 1 L/min for fouling control and microalgae mixing purpose. Water from the tank was extracted through the membrane using a peristaltic pump. The membranes were washed with sodium hypochlorite (NaOCl, John Kollin) once the permeation flux dropped to 5 mL/m^2·h. The average flux of 150 h filtration with 5-min interval recording was used as the analysis input. Hence, the data accuracy should be reliable and acceptable (Stieberova et al., 2017; Zuin et al., 2013).

2.2. LCA methodology

LCA with Simapro software is widely used to quantify the impact of a product towards environment throughout its life cycle from the production of raw materials to its final disposal. There are four phases included in the LCA analysis: goal and scope definition, inventory, life cycle impact assessment and data interpretation. Firstly, the objectives and the boundaries of this study were determined. The functional unit of the products which was used as a comparison basis was defined. The LCA analysis could be "cradle-to-grave" or "cradle-to-gate", which may or may not include recycling. In the second phase, the inputs and outputs information (foreground and background data) involved in the products life cycle were connected to the subjects under investigation. The impacts of the products' life cycle towards environment were quantified in the third phase using various life cycle impact assessment (LCIA) methods. Lastly, the outcomes of the LCIA were interpreted in details. In this study, Simapro 8.4.0 was used as the LCA analysis tool.

2.2.1. Goal, functional unit and system boundaries

The main objective of this study was to compare the environmental impact of two A-MPR system operated with different sets of membranes: Ag/GO-PVDF membrane and neat PVDF membrane. As the membranes used in the system were not available in the Simapro database, the information of membrane fabrications were created and the results were compared. The functional unit for membrane fabrication was defined as "1 g of membrane". The functional unit for A-MPR operation was "1 L of permeate", where the water was biologically treated by microalgae in the A-MPR tank followed by membrane filtration. Therefore, all the raw material inputs, energy consumption and emissions, as well as impacts calculated in the software were based on the functional unit of 1 g membrane and 1 L permeate.

The system boundaries of the A-MPR unit is illustrated in Fig. 2. The components and activities outside the dotted line were not considered in the LCA. This study is a cradle-to-gate approach where the construction of the setup started from raw materials to the produce of permeate. The disposal or reuse of the materials were excluded. The equipment such as tank, pump and air compressor might be reused for other laboratory work. They might be recycled if they are broken, however, the recycling activities is not fully practised in the country. The permeate was used for washing of glassware in the laboratory where the used water was drained into the municipal wastewater piping system. Microalgal biomass harvested from the A-MPR tank was not included. Many assumptions were to make if all the disposal routes were taken into consideration which will greatly affect the applicability/relevance of the results. The lifespan of the system is 20 years according to previous literature (Ioannou-Ttofa et al., 2016).

2.2.2. Inventory data and limitations

The raw materials input, energy consumption, transportation and emissions in different forms involved in the construction and operation of the A-MPR unit were collected and categorized. The life cycle inventory of the A-MPR unit is shown in Table A2. Most LCA data were taken from the Simapro database. However, some assumptions were made when the required data could not be found in the database:

- The PVDF was replaced by 50% tetrafluoroethylene (TFE) and 50% polyethylene (PE), which was confirmed by Solvay representatives as acceptable for its environmental influence (Zackrisson et al., 2010).
- Main materials of the manufacturing equipment were selected as the construction data. ABS and steel were considered as the main materials for pump, and cast iron was selected for air compressor. The lifetime of pump and air compressor were determined according to the suppliers' advice.
- The inventory data for synthesis of Ag/GO nanohybrids, the production of Ag/GO-PVDF membrane and neat PVDF membrane in laboratory scale were not available in Simapro and previous literature. Therefore, the primary data were established by reviewing the whole process (Chong et al., 2017a,b) in our laboratory. The life cycle inventory of Ag/GO nanohybrids synthesis and membrane fabrication are presented in Table A3 and A4, respectively. PVP as membrane pore forming agent was not included due to the little amount which may give insignificant environmental impact and the data is unavailable in Simapro database (Al-Sarkal and Arafat, 2013). Please note

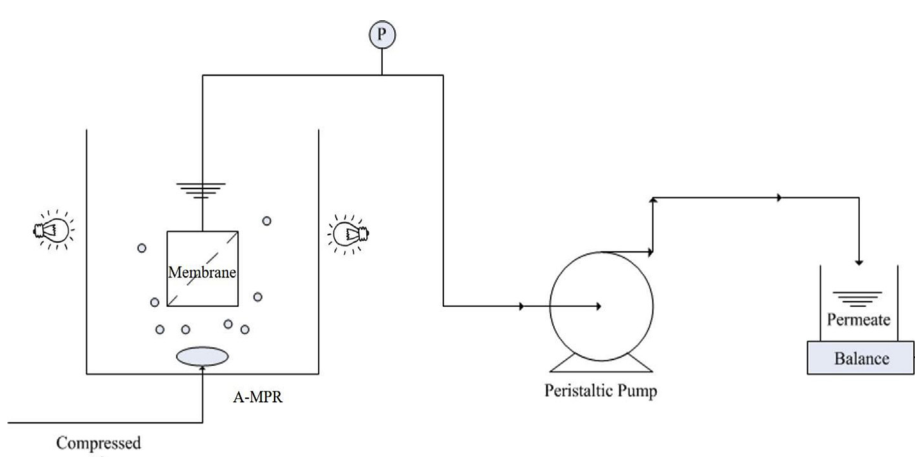

Fig. 1. Schematic diagram of A-MPR.

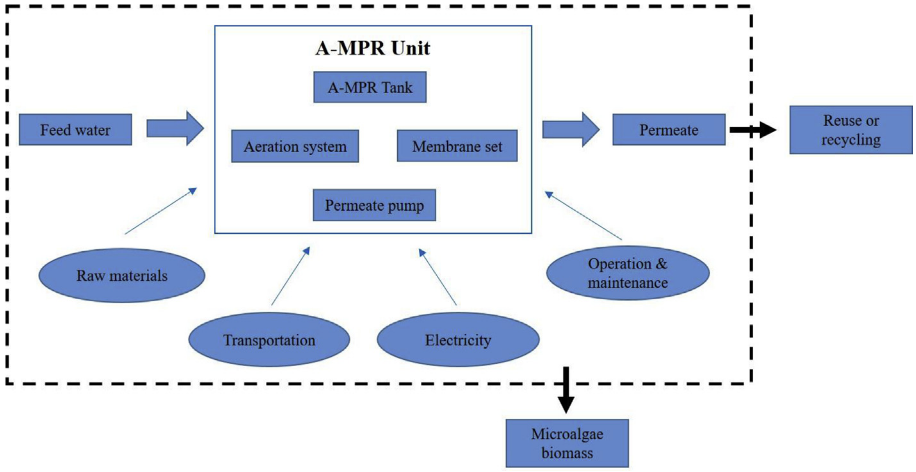

Fig. 2. System boundaries of A-MPR unit for LCA analysis.

that the inventory data did not include synthesis of Ag/GO nanohybrids for neat PVDF membrane fabrication. Construction of equipment was excluded in synthesis of Ag/GO nanohybrids and membrane fabrication process.

- The lifetime of both PVDF membranes was assumed to be 5 years, which was similar to the lifetime of commercial membranes. Our previous study had proven that there was no leaching of the nanomaterials from the PVDF membrane (Chong et al., 2017a).

2.2.3. Life cycle impact assessment

ReCiPe 1.13 (H) and IPCC 2013 GWP 100a as life cycle impact assessment methods were selected in this study. ReCiPe 1.13 (H) was used to indicate impacts of the activities in a broad set of environmental issues. ReCiPe combined both midpoint and endpoint approaches. There were 18 impacts included in midpoint: climate change (CC), ozone depletion (OD), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (MEP), human toxicity (HT), photochemical oxidant formation (POF), particulate matter formation (PMF), terrestrial ecotoxicity (TET), freshwater ecotoxicity (FET), marine ecotoxicity (MET), ionizing radiation (IR), agricultural land occupation (ALO), urban land occupation (ULO), natural land transformation (NLT), water depletion (WD), metal depletion (MD) and fossil depletion (FD). These could be further classified into 3 endpoints categories, which were human health, ecosystem and resources. IPCC 2013 GWP 100a was selected to present the global warming potential (GWP) of the involved activities in terms of CO_2 emission equivalent ($CO_{2\text{-eq}}$) with 100 years' time frame. This approach is normally used in reporting as it is understandable by the public. All the analyses conducted are generally based on Malaysia grid in Simapro unless stated specifically.

3. Results and discussion

3.1. AG/GO-PVDF and PVDF membranes fabrication

Fig. 3a shows the impact assessment of the Ag/GO-PVDF membrane and PVDF membrane fabrication towards environment using ReCiPe midpoint analysis. As explained in Section 2.2.3, a total of 18 impacts categories were included in the midpoint indicators results. The value of each impact category was listed in Table A5. From the results, the environmental effects of the Ag/GO-PVDF membrane fabrication were higher than the PVDF membrane in almost all of the impact categories due to the additional synthesis process of Ag/GO nanohybrids. Among the impact categories, metal depletion showed the greatest difference between the two membranes. The indicator results for Ag/GO-PVDF and PVDF membranes were 8.69×10^{-4} kg Fe eq./g and 2.39×10^{-4} kg Fe eq./g, respectively. Silver nitrate ($AgNO_3$) as one of the precursor to synthesize Ag/GO nanohybrids was produced by reacting silver (Ag) with nitric acid. Therefore, taking Ag from the nature resulted in metal depletion. On the other hand, both of the membrane fabrication processes gave similar effect (100%) to ozone depletion as same amount of PVDF polymer were used for the casting solution preparation. Further analysis on the influence of each material used in Ag/GO-PVDF membrane fabrication towards the impact categories were illustrated in Fig. 3b. The results revealed that PVDF polymer was the only factor that contributed to the ozone depletion. From the analysis, an amount of 3.84×10^{-6} kg of CFC-11 eq./g of membrane (ozone depleting substances) was emitted into the atmosphere, contributed by the polymerization of PVDF in chlorofluorocarbon (CFC) solvents during its manufacturing process (McKeen, 2016). The findings was similar to previous studies performed by Zuin et al. (2013) and Stieberova et al. (2017), where PES and PVDF polymers contributed significant effects on global warming.

According to the results shown in Fig. 3c, the significance of environmental impact of membrane fabrication was evident for CC, OD, TA, FE, HT, PMF, FET, MET and FD. Climate change (CC) and human toxicity (HT) were contributed by the manufacturing of PVDF polymer and electricity used during the membrane fabrication process. The electricity used was the main contributor to almost all of these impact categories. The electricity generation in Malaysia is heavily dependent on burning fossil fuel with 48% of coal and 46% of natural gas; while only 5% of the energy comes from hydro power and 2% from renewable energy (ST, 2016). This was supported by the results in Fig. 3b where CC, TA, FE, HT, PMF, FET, MET and FD showed high values due to fossil fuel usage and their figures remained high after normalization as shown in Fig. 3c. Besides, it can be concluded that the addition of Ag/GO nanohybrids into PVDF polymeric membrane did not give significant effect on most of the impact categories, but has mainly influenced the marine ecotoxicity and metal depletion due to mining activities (Fig. 3b). This was because only a little amount (0.4%) of the nanohybrids was required (blended into the PVDF polymeric solution) to give remarkable improvement on the membrane performance. On the other hand, the membranes showed little impacts on MEP, POF, TET, IR, ALO, ULO, NLT, WD and MD impact categories.

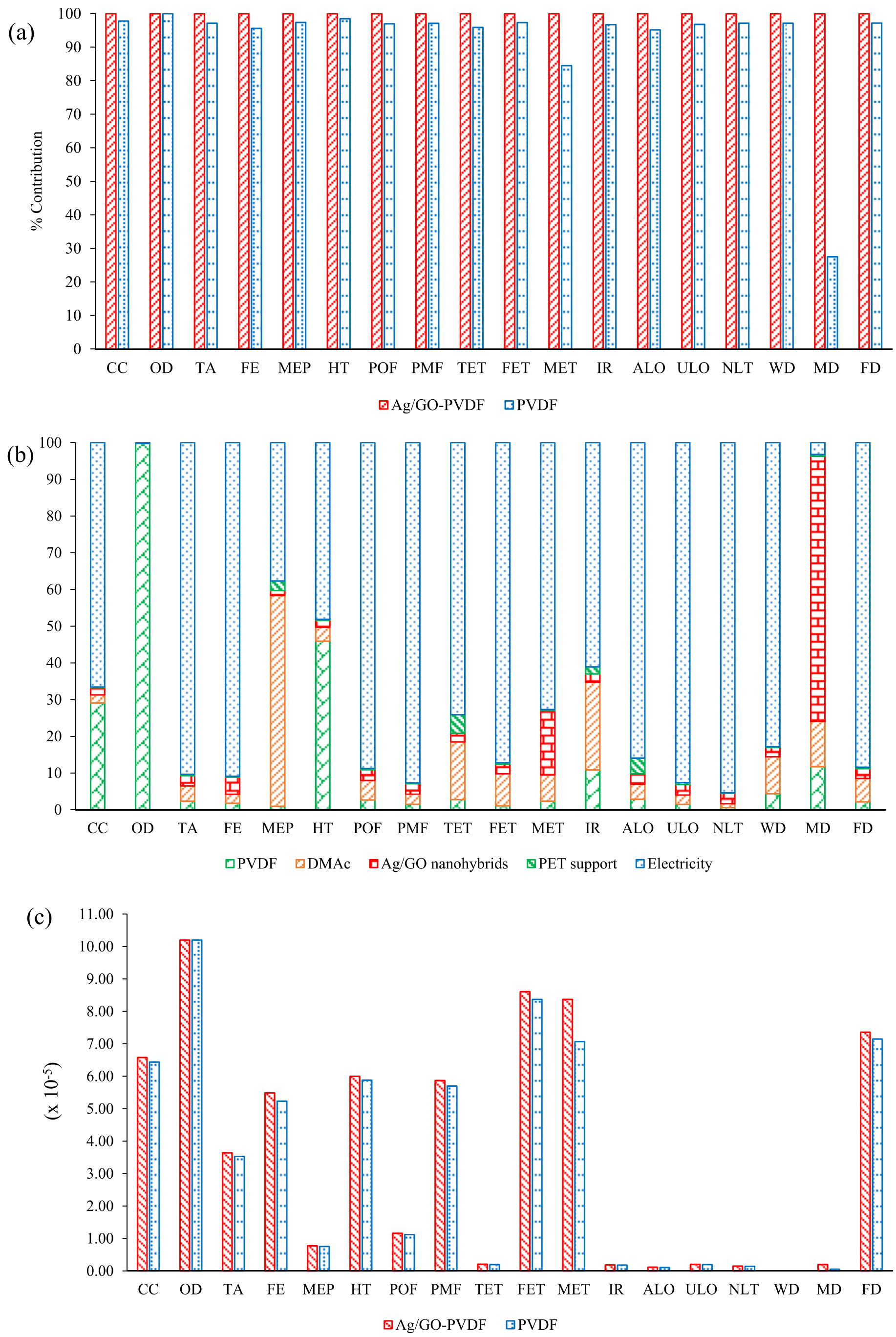

Fig. 3. (a) Comparison of midpoint level, (b) contribution of each parameter, and (c) normalized midpoint level on environmental impacts of Ag/GO-PVDF and PVDF membranes fabrication using ReCiPe. *Analysis based on Malaysia grid in Simapro.

3.2. Comparison of Ag/GO-PVDF and PVDF membranes in A-MPR operation

3.2.1. Midpoint analysis

The effects of membrane type on the midpoint and endpoint's environmental impact categories in the A-MPRs' operation was further identified by employing ReCiPe assessment method. The operation of A-MPR with the Ag/GO-PVDF membrane was able to reduce all of the environmental impact categories in similar ratio as presented in Fig. 4a, hence improving the overall system life cycle. The incorporation of the Ag/GO nanohybrids in the PVDF membrane had improved the hydrophilicity of the membrane, resulted in higher volume of permeate as the system output (Chong et al., 2017b). The average output of the permeate for the Ag/GO-PVDF membrane and PVDF membrane were 6.89 L/m^2·h and 5.79 L/m^2·h, respectively. Therefore, the A-MPR with Ag/GO-PVDF membrane gave lower values of inputs when rationalizing the input figures with functional unit of 1 L. For instance, the Ag/GO-PVDF membrane had higher impact in metal depletion category compared with the PVDF membrane in fabrication process (Fig. 3a). However, the latter showed higher impact during the operation of A-MPR (Fig. 4b). Furthermore, the Ag/GO-PVDF membrane possessed high biofouling resistance, which remarkably reduced the attachment and growth of bacteria on membrane surface

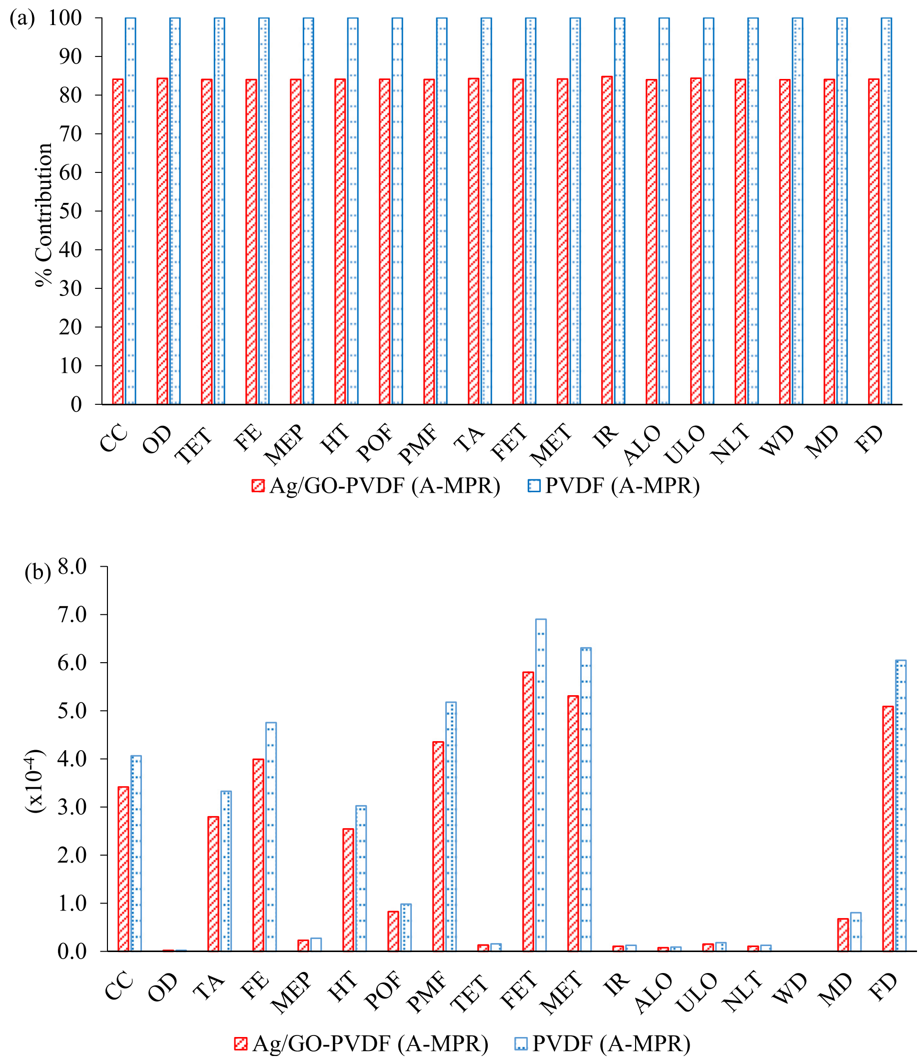

Fig. 4. (a) Comparison of midpoint level and (b) normalized midpoint level, on environmental impacts of Ag/GO-PVDF and PVDF membranes in A-MPR using ReCiPe. *Analysis based on Malaysia grid in Simapro.

(Mahmoudi et al., 2015). Hence, it has greatly reduced the amount of membrane cleaning agent (NaOCl), which was used in this study. The figure also showed that the usage of the Ag/GO-PVDF membrane did not impose significant effect on a specific category, probably due to the little amount of Ag/GO nanohybrids incorporated into the membrane.

On the other hand, Fig. 4b presents the normalized results from Fig. 4a, showing both of the A-MPR operations gave the highest impact on FET, followed by MET and FD. Other categories with relatively high impact were CC, TA, FE, HT and PMF. Detailed analysis of the parameters that contributed to each category was identified and illustrated in Fig. 5. Permeate pump had the highest influence on most of the impact categories, followed by air compressor, weighing balance and piping. Coincidently, the inputs of these equipment's power consumption followed the above impact sequence. This implied that the electricity usage of these equipment had the most significant effect on the environment during the A-MPR operation. The energy source in the country is highly dependent on fossil fuel (burning of coal and natural gas). These fossil fuels released heavy metals (nickel, beryllium, copper and etc.), sulfuric compounds and polycyclic aromatic hydrocarbons (PAHs) into the environment by outflowing into nearby river or leaching into the soil during the extraction and waste disposal process (Atilgan and Azapagic, 2015). Therefore, these activities imposed great effect on MET and FET categories. Climate change was mainly contributed by the release of methane (CH_4) during coal mining and natural gas extraction as well as emission of carbon dioxide (CO_2) during combustion of these fossil fuel (Chiari and Zecca, 2011; Höök and Tang, 2013). Coal fired stations emit about 30 billion tons of CO_2 per year worldwide (Brook et al., 2014). The release of these gaseous have been known as the major culprits that lead to global warming. Besides, particulate matter, heavy metal, mercury, sulfur dioxide (SO_2), nitrogen oxides (NO_x) and carbon monoxide (CO) released from the coal-fired and natural gas power plant formed smog and acid rain, contributing to TA, FE, POF and PMF impact categories. The acidifying N and S deposition on the forest had led to soil acidification and subsequently loss of some plant species (Azevedo et al., 2013; Soons et al., 2017). On the other hand, the major contributors of human toxicity were due to generation of heavy metal such as selenium, molybdenum, beryllium and barium during coal combustion. Emission of chromium, arsenic and nickel from plant construction was the largest contributor, followed by gas extraction (Atilgan and Azapagic, 2015). According to Fig. 5, transportation in this study was associated with many of the impact categories: CC, OD, HT, POF, TET, MET, IR, ULO and FD. Malaysia is blessed with abundance of natural resources and citizen

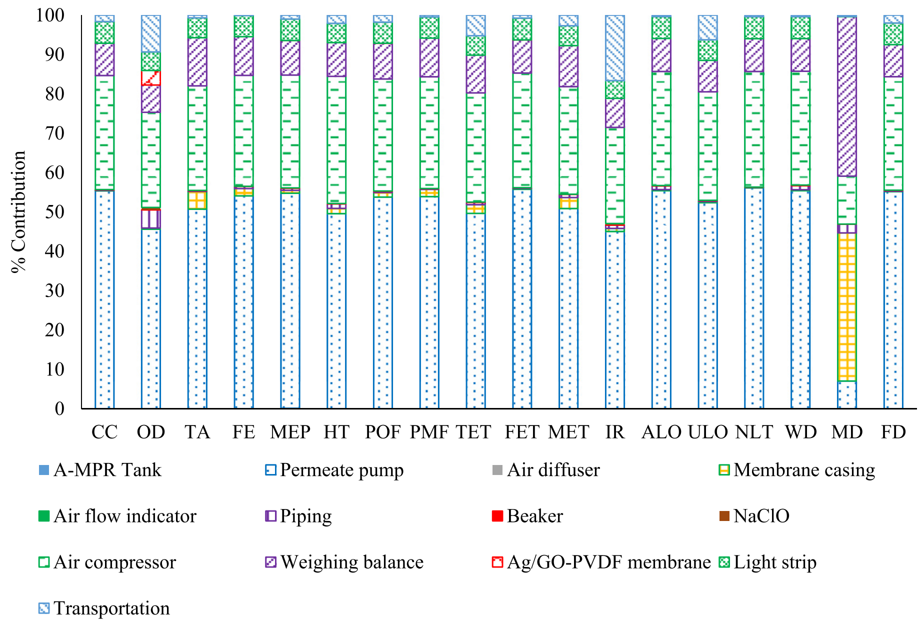

Fig. 5. Contribution of each parameter of A-MPR (using Ag/GO-PVDF membrane) on midpoint impact categories using ReCiPe. *Analysis based on Malaysia grid in Simapro.

are enjoying petrol, diesel and natural gas with relatively low price as the main fuel for transportation. According to a report by Briggs and Leong (2016), almost all motorcycles and passenger cars run on petrol. The fuel usage of new registered motor vehicles was 94.12% petrol, 4.82% diesel, 0.6% petrol and electric, 0.39% petrol and natural gas, 0.05% electric, and 0.01% NGV. Therefore, the CO_2 emission in the country contributed by road vehicles was shown to be very high (85.3%) in a study by Hosseini et al. (2013). The CO, PM especially below 2.5 μm ($PM_{2.5}$) and other toxin gaseous emitted from vehicles imposed high impact on the environment. The concentration of $PM_{2.5}$ elevated along the roadway and inhalation of these $PM_{2.5}$ results in a broad range of negative health effect (Gan et al., 2010; Poorfakhraei et al., 2017). Other parameters exhibited relatively small impact towards the environment. Although the membranes showed some scores in the OD category in Fig. 5, its influence on the overall A-MPR operation was minimum (Fig. 4b). Besides, the membrane casing was made from stainless steel, therefore it mainly contributed to MD impact category. Meanwhile, polyvinyl chloride (PVC), silicone and brass were the materials used for valves and tubing of the system. Therefore, these materials gave impacts on ozone depletion and metal depletion. Beaker, A-MPR tank, air flow indicator, air diffuser and NaOCl exhibited almost negligible effect on all of the impact categories. The high consumption of NaOCl on PVDF membrane cleaning (consumed one fold more than Ag/GO-PVDF membrane) might have increased the operational cost of the system, but did not have obvious impact on the environment. On the other hand, the overall A-MPRs' operation imposed only little effects on the following impact categories (Fig. 4b): OD, MEP, TET, IR, ALO, ULO, NLT and WD.

3.2.2. Endpoint analysis

Fig. 6 presents the results of ReCiPes' three endpoint indicators which consists of human health, ecosystem and resources. The A-MPR with the Ag/GO-PVDF membrane showed less effects on all of the three impact categories compared with the A-MPR with PVDF membrane. The total environmental footprint of the A-MPR with PVDF membrane was 290 mPt per L permeate. On the other hand, the environmental footprint of A-MPR with the Ag/GO-PVDF membrane has reduced by 15.9%, to 244 mPt per L permeate. This

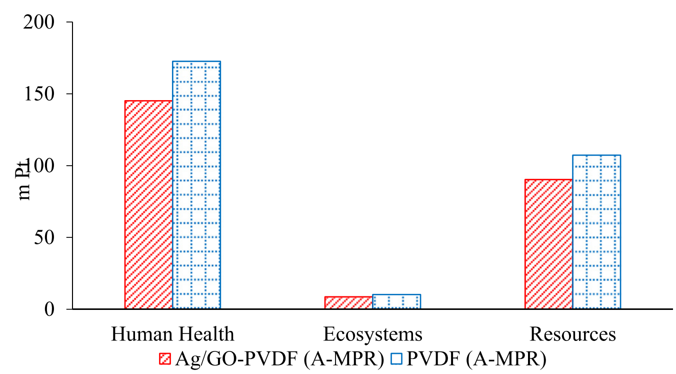

Fig. 6. Endpoint impact categories of A-MPR (Ag/GO-PVDF and PVDF membranes) using ReCiPe. **Analysis based on Malaysia grid in Simapro.*

indicated that the incorporation of the nanohybrids into the membrane was more environmental-friendly as it showed encouraging results in reducing the environmental impacts. From the figure, human health exhibited the highest impact, followed by resources and ecosystems for both of the A-MPR systems. These findings was similar to other studies by Pretel et al. (2013) and Ioannou-Ttofa et al. (2016). The results could be interpreted as the system affected human health the greatest, due to the use of fossil fuel as the main source for energy generation. The extraction, processing, burning and waste disposal of these fossil fuel deteriorated the environment in various forms (i.e. release of greenhouse gases, particulate matter and toxic substances). These pollutants may come into contact with human directly or indirectly via inhalation, food intake or skin contact. On the other hand, the continuous extraction of fossil fuel would definitely lead to resources depletion as coal, natural gas and petroleum are non-renewable energy sources on planet earth.

3.3. Sensitivity analysis

Based on the above study, one could identify that electricity consumption from combustion of fossil fuel was the main

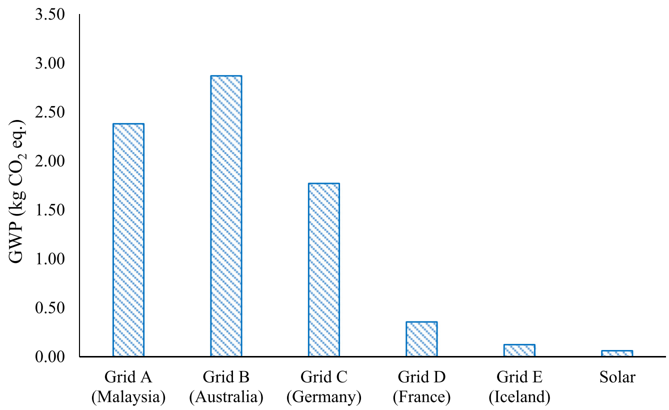

Fig. 7. Global Warming Potential for 1 L of permeate produced by A-MPR.

environmental hotspot that affect the overall sustainability of the A-MPR system. Malaysia is one of the countries that highly rely on fossil fuel especially coal and natural gas as energy sources. Reducing the burning of fossil fuel could certainly alleviate the emission of greenhouse gases (GHG) or other toxic substances. Sensitivity analysis was performed to evaluate the significance of energy sources, Grid A (Malaysia), Grid B (Australia), Grid C (Germany), Grid D (France), Grid E (Iceland) and solar photovoltaic on sustainability of the A-MPR system. The energy source of Malaysia and Australia are quite similar as both are highly dependent on coal and natural gas. Australia, which is the main producer and exporter of coal, generates around 63% of its energy from coal burning, 21% of natural gas and 14% of renewable energy (DoEE, 2017). Electricity mix of France consists mainly of nuclear power (78%), the remaining are renewable energy (16%) and fossil fuel (6%) (IEA, 2017). The amount of renewable energy supply (33%) in Germany is higher than nuclear energy (15%), and the rest are coal (43%), and natural gas (9%) (Frunhofer ISE, 2016). Iceland is known as one of the countries that utilizes almost 100% of renewable energy, consists of 73% hydropower and 27% geothermal (NEA, 2016). Besides, Malaysia as a tropical country, blessed with abundance of sunshine throughout the year has high potential for solar photovoltaics energy generation. Therefore, solar photovoltaic was selected for the sensitivity analysis.

Fig. 7 shows the global warming potential (GWP) of different electricity mix. The greenhouse gases emitted by Grid B was the highest among the electricity mixes with 2.98 kg CO_2 eq./L. The emission was 20.6% higher than Grid A although it had higher contribution from renewable energy. This was because Grid B had high reliance on coal burning that severely deteriorated the environment as discussed in Section 3.2.1. Meanwhile Grid A utilized higher quantity of natural gas which was known to be the cleanest fuel among the fossil fuels (Liang et al., 2012; Xu and Chen, 2016). Grid C's emission was 2.0 kg CO_2 eq./L which is 25.6% lower than Grid A. Germany was once relied highly on fossil fuel but now is shifting its direction towards greener energy sources. The country has been aggressively investing on renewable energy harvesting technology especially on solar photovoltaic in these recent years. Besides, the nuclear power in the country is relatively low compared with other European countries. Grid D which is heavily dependent on nuclear energy emitted only 0.417 kg CO_2 eq./L to the environment. The emission dropped sharply by 94.8% compared with conventional electricity mix (Grid A). Nuclear energy imposed relatively low impact on global warming as the technology cutting down massive amount of CO_2 and other greenhouse gases. The nuclear power plants in the world have prevented the emission of 2 billion tons of CO_2 annually (Brook et al., 2014). Grid E's emission was the lowest among the grids due to 100% renewable energy generated from hydropower and geothermal. The amount of wind power and fossil fuel was less than 0.001% which was insignificant in the total electricity generated (NEA, 2016). Lastly, if solar photovoltaic is fully implemented in Malaysia, the emission could be reduced by 97.5%.

The impact of different electricity mixes on environmental damage categories is shown in Fig. 8. The data were also presented in Table A6. Overall, Grid A, Grid B and Grid C showed high impact on most of the categories: CC, TA, FE, HT, PMF, FET, MET and FD which was due to the relatively high usage of fossil fuels especially coal. Compare with natural gas-fired plants, coal-fired power stations emitted 50% more CO_2, 70% more NO_x and 99% more SO_2 (Xu and Chen, 2016). Coal is widely used as main fossil fuels for electricity generation in developing countries as it is more economic and easy to handle due to the flexible transportation system where it can be delivered with trucks, trains and ships (Liang et al., 2012). Results shows that ionizing radiation (IR) was the prominent concern for Grid D which was dominated by nuclear energy, while the scores in other categories were relatively low. The process of mining, washing and transportation of uranium contributed to TA, FE, HT, PMF, FET, MET, MD and FD impact categories (Ferrari et al., 2017). However, the operation of nuclear power plant did not produce air pollution or gas CO_2. On the other hand, Grid E and Grid

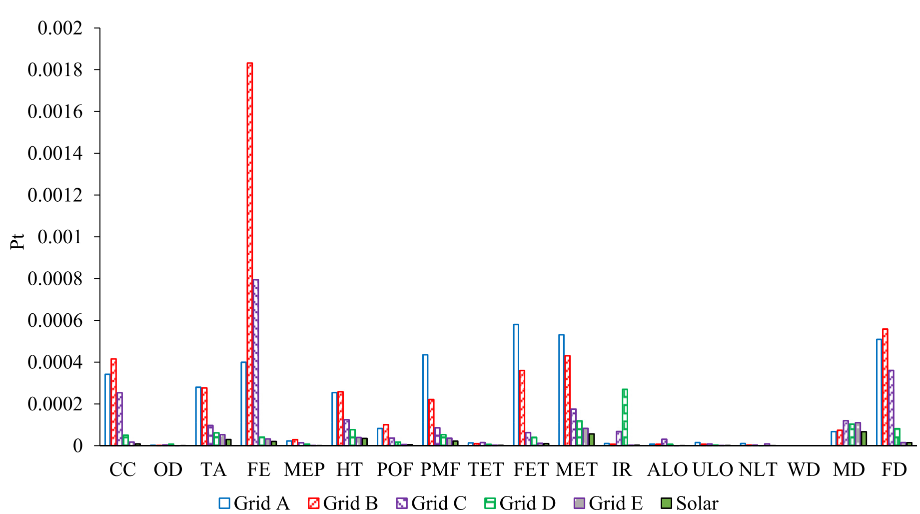

Fig. 8. ReCiPe's normalized results for A-MPR with different grids.

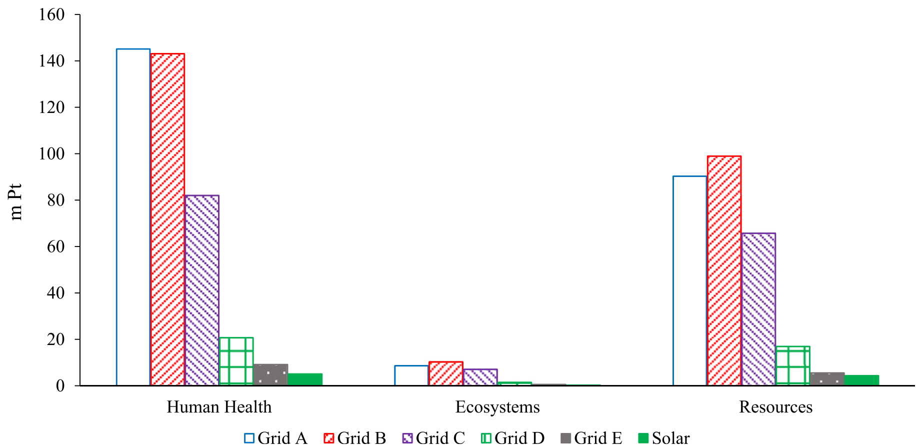

Fig. 9. Endpoint impact categories of A-MPR with different grids.

F show low scores in the assessment and negligible in most of the impact categories. As observed in Fig. 9, obviously the use of energy source paid a great influence on the environment. The scores of Grid A, Grid B and Grid C were relatively high in human health and resources categories. Meanwhile, the impact of Grid E and Grid F were almost negligible in all the three impact areas. This imply that a sustainable A-MPR system can be developed with the use of renewable energy (hydropower, geothermal and solar photovoltaic). Therefore, the shifting of fossil fuel towards renewable energy with cost effective technology is critical in preserving the environment while enjoying the economy benefit.

4. Conclusions

The modified PVDF membrane with Ag/GO nanohybrids did not impose significant impact on a specific damage category but affected the overall system life cycle in a positive way, as the output of the system was greatly improved with higher membrane permeability and reduced membrane fouling. Although the overall environmental footprint of the Ag/GO-PVDF membrane fabrication process was higher, its application in the A-MPR outweighed its fabrication life cycle's results due to higher throughput in the A-MPR operation. This indicated that the application of the Ag/GO-PVDF membrane in A-MPR system was more sustainable in long-term operation. Besides, the application of NaOCl as cleaning agent showed very little influence on the environment compared to electricity consumption. The high usage of coal as fossil fuel in electricity generation (midpoint analysis) was the main environmental hotspot which has greatly affected human toxicity (endpoint analysis). Energy demand as the crucial parameter in the LCA analysis showed impressive improvement when the current electricity mix was replaced with renewable energy. More effects shall be devoted in shifting the current electricity mix towards renewable energy such as fuel cell, solar photovoltaic, hydropower and windpower in preventing environmental pollution while enjoying the economic benefit.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2018.08.121.

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